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<p>(54) Title: <b>ONE COMPONENT, MOISTURE CURABLE INSULATION ADHESIVE</b></p> <p>(57) Abstract</p> <p>A one-component, moisture-curable adhesive composition is provided for securing insulation to a roof deck. The adhesive comprises a silylated polymer and an extender. The adhesive composition may also include a plasticizer, a moisture scavenger, and a catalyst. Upon curing, the adhesive composition exhibits an uplift pull strength of at least about 200 Lbs/ft<sup>2</sup>.</p>			

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## ONE COMPONENT, MOISTURE CURABLE INSULATION ADHESIVE

The present invention relates to a one component, moisture curable adhesive for use in securing roofing insulation to a roof deck, and more particularly, to a 100%  
5 solids, solvent free adhesive containing a silylated polymer.

Adhesives are known in the art for bonding insulation to roof decks used in  
roofing and reroofing applications, particularly "flat roofing". By "flat roof", it is meant a  
10 roof having a slope of less than about 25° relative to a horizontal plane. Such roofs  
typically comprise a waterproof membrane, a thermal insulation layer, and the roof  
deck. Two common methods of securing insulation to roof decks include the use of  
15 mechanical fasteners and hot applied asphalt.

The use of mechanical fasteners is very labor intensive. For example, roofs  
with concrete decks often require a pilot hole to be drilled before the fastener can be  
anchored. On concrete decks, particularly older structures, the pilot holes may cause  
15 cracks to form in the deck, compromising deck integrity.

In addition, while many fasteners are protected with anti-corrosive coatings,  
corrosion can still occur. The protective coating can be scratched when ratcheting  
fasteners into place. Minor nicks in the coating can be enough to allow corrosion to  
penetrate the entire fastener. Even where fasteners do not corrode, they will  
20 generally expand and contract with changes in weather conditions. Therefore, holes  
through which fasteners are anchored are prone to enlargement over time, reducing  
holding strength and eventually allowing the fastener to back out through the  
waterproofing membrane.

The use of hot asphalt has also been widely used to secure insulation to roof  
25 decks; however, the use of asphalt requires the use of a heating kettle which often  
requires special equipment for transport to the roof top, adding to the cost of the job.  
In addition, some light-weight decks are not capable of supporting such a load.  
Further, as most heating kettles in use are heated by flames, a potential fire hazard  
exists. The hot adhesive also reaches temperatures in the range of 300 to 500°F  
30 (148 to 260°C), posing a potential hazard to those working with the material, and  
also produces offensive fumes. Further, hot asphalt tends to have a rapid cool off  
rate, especially during cold weather conditions. Accordingly, the insulation must be

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applied immediately to the asphalt or it will lose its ability to wet out the insulation and bond the insulation to the deck.

Accordingly, there is still a need in the art for an adhesive which may be used to secure insulation to roof decking without the need for mechanical fasteners or hot applied asphalt.

The present invention meets that need by providing a one-component, moisture curable, solvent-free adhesive for use in securing insulation to roof decks.

According to one aspect of the present invention, a one-component, moisture-curable adhesive composition for securing insulation to a roof deck is provided comprising a silylated polymer selected from the group consisting of silylated polyurethanes and silylated polyethers, and an extender selected from the group consisting of coal tar, aromatic oils, and hydrocarbon resins. By "one-component" adhesive, it is meant that the adhesive is ready for use as removed from its packaging as it contains enough reactive components to cure it under ambient conditions. The extender preferably comprises coal tar.

The adhesive composition preferably further includes a plasticizer selected from the group consisting of phthalates, mellitates, benzoates, phosphates, butyrates, polymeric esters, adipates, sebacates, aromatic and cyclic oils, epoxidized oils, and solvents. Preferably, the plasticizer comprises diundecyl phthalate.

The adhesive composition also preferably comprises a moisture scavenger. A preferred moisture scavenger is 4,4,-diphenylmethane diisocyanate.

The adhesive composition also preferably includes at least one catalyst selected from the group consisting of dibutyl tin diacetate, 1,8-diaza-bicyclo (5.4.0) undecene-7, (bis(2-dimethylaminoethyl)ether, and combinations thereof.

In a preferred embodiment of the invention, the adhesive composition comprises from about 20 to 50% by weight of a silylated polymer, from about 30 to 75% by weight of an extender, from about 2 to 25% by weight of a plasticizer, and from about 0.5 to 10% by weight of a moisture scavenger. The silylated polymer is selected from the group consisting of silylated polyurethanes and silylated polyethers.

The roofing insulation is preferably adhered to a roof deck by applying the adhesive composition to a roof deck substrate, placing the insulation in contact with

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the adhesive composition on the roof deck substrate, and allowing the adhesive composition to cure. Upon full cure, the adhesive composition exhibits an uplift pull strength of at least about 200 lbs/ft<sup>2</sup> (97 g/cm<sup>2</sup>).

Accordingly, it is a feature of the present invention to provide an adhesive 5 composition for adhering insulation to roofing deck without the need for mechanical fasteners or hot applied asphalt. Other features and advantages of the invention will be apparent from the following description, the accompanying drawings, and the appended claims.

Fig. 1 is a perspective view of a roof construction comprising roofing insulation 10 adhered to a roof deck substrate with the adhesive composition of the present invention.

The adhesive composition of the present invention provides many advantages over prior art methods of adhering roofing insulation to a roof deck. First, the use of silylated polymers broadens the choice of materials that can be used in the adhesive 15 formulation as opposed to more limited choices available when using conventional polymers containing active isocyanate groups. Such conventional polymers react with and limit the stability of compounds containing active hydrogen atoms. Silylated polymers are more desirable because they cure selectively upon exposure to moisture and are stable in the presence of other active hydrogen atoms. For example, 20 silylated polymers form stable compounds when blended with coal tar extenders which have active hydrogen atoms, while conventional polymers react with the active hydrogen atoms.

The silylated polurethane of the present invention may be made by the process described in U.S. Patent No. 3,632,557, the disclosure of which is hereby 25 incorporated by reference. Another preferred method for making silylated polyurethanes and polyethers is described in commonly assigned copending application Serial No. 09/218,573 entitled "FAST-CURE SILYLATED POLYMER ADHESIVE", the disclosure of which is hereby incorporated by reference. A preferred silylated polyether is also commercially available from Kaneka under the 30 designation MS polymer.

A coal tar extender, or filler, is preferably included in the composition which functions to increase adhesion through improved wetting and aggressive tack.

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Preferred coal tar extenders for use in the present invention include RT-5, RT-7, RT-12, CP 250 and CP 524, all commercially available from AlliedSignal, Inc. Other suitable coal tar extenders include RT-6, RT-7, and RT-8, all commercially available from Crowley Chemical Company. Such coal tars generally comprise a mixture of 5 organic compounds including 1-40 ringed polynuclear aromatic hydrocarbons and some substituted compounds. The coal tars are identified by grades set forth by ASTM requirement D490.

Suitable alternative extenders include, but are not limited to, aromatic oils and hydrocarbon resins. Suitable hydrocarbon resins include, but are not limited to, rosin esters, aromatic resins, and polyterpene resins, all commercially available from 10 Hercules under the respective designations Stabelite Ester 10, Piccovar AP-10 and AP-25, and Piccolyte S 25; cumarone indene and modified cumarone indene resins available from Neville Chemical Company under the designations Cumar R-27, R-29, P-10, P-25 and FT 4-9; aromatic petroleum hydrocarbon resins available from Neville 15 Chemical Company under the designations Nebony L-55 and L-100; rosin oil/tall oil resins available from Sovereign Chemical Company under the designations Tartac 20, 30 and 40; and polyterpene and rosin resins available from Arizona Chemical Company under the designations Zonarez Alpha 25 and Zonester 25, respectively.

Suitable aromatic oils include, but are not limited to, Sundex 790 and 8125, 20 Sundex T 840, 750T, 780T, 790T, 7110T and 8600T, all available from Sun Petroleum Products Company; aromatic oils and plasticizers available from Neville Chemical Company under the designations NP-10, NP-25, CB-4-34, X-743 and 745; and aromatic oils available from Exxon Chemical Company under the designations Flexon 340 and 391, and Telura 126 and 171.

25 The adhesive composition also includes a moisture scavenger to scavenge moisture from the composition. A preferred moisture scavenger is 4,4-diphenylmethane diisocyanate (MDI), commercially available from Dow Plastics under the designation Isonate 50-OP. Other suitable moisture scavengers include molecular sieves such as 3A, 4A, 5A and 13X from UOP, lime (CaO) from Mississippi 30 Lime Company, organosilanes, and isocyanates having molecular weights less than 500.

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A catalyst is preferably present to increase the curing reaction rate of the silylated polymer upon exposure to moisture. The curing reaction is believed to take place with the hydrolyzable alkoxy groups being replaced with hydroxyl groups.

Crosslinking takes place through the condensation of pairs of SiOH groups to siloxane linkages (Si-O-Si) or by the reaction of SiOH groups with SiO-alkoxy groups to form a siloxane linkage and an alcohol. Catalysts useful in the present invention are known in the art and include, for example, tin compounds, alkyl titanates, organosilicon titanates, metal salts of carboxylic acids such as stannous octoate, dibutyltin diluariate and the like, amine salts such as dibutylamine-2-ethylhexoate and the like, or other conventional acidic and basic catalysts. A preferred catalyst is 10 stanne,bis(acetoxy)dibutyl, commercially available from Air Products under the designation METACURE™ T-1. Another suitable tin catalyst is 1,8-diaza-bicyclo (5.4.0) undec-7-ene, commercially available from Air Products under the designation POLYCAT® DBU. Still other suitable catalysts include amine catalysts, such as 15 (bis(2-dimethylaminoethyl)ether, commercially available from Air Products under the designation DABCO BL-19.

A plasticizer is preferably contained in the adhesive composition to extend the viscosity range and control the cohesive strength level. Preferred plasticizers include phthalates such as diundecyl phthalate ( DUP), mellitates, benzoates, phosphates, 20 butyrates, esters, adipates, sebacates, aromatic and cyclic oils, epoxidized oils, and solvents such as toluene, xylene, and methanol.

Various other additives may be added to modify the physical properties of the composition, for example, fillers, e.g., reinforcing fillers, such as fume silicas, silica aerogels and precipitated silicas of high surface area. Non-reinforcing fillers can also 25 be used, for example, coarse silicas, such as diatomaceous earth, crushed quartz or metallic oxides, such as titania, ferric oxide, zinc oxide, talc and the like. In addition, fibrous fillers such as glass fibers or filaments may be used. In all cases where filler is used, it is desirable that the filler be substantially dry before it is admixed with the silylated polymer. The fillers are generally used to upgrade physical properties and to 30 modify the flow characteristics of the uncured adhesive.

The adhesive composition may also contain additives such as pigments, UV stabilizers, oxidation inhibitors and the like, or fillers/pigments such as graphite and

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carbon black. Other possible additives include flame retardants, blowing agents, perfumants, antistats, insecticides, bacteriostats, fungicides, surfactants, and the like.

The components of the adhesive composition are preferably mixed until homogenous and then packaged as desired. It should be appreciated that the mixing 5 and packaging of the adhesive composition must be done under anhydrous conditions. This is preferably achieved by use of nitrogen purge in mixers and holding tanks and by use of nitrogen purge and blanketing during packaging.

Referring now to Fig. 1, a roofing system 10 in accordance with the present invention is illustrated. The roofing system includes a roof deck 12, roofing insulation 10, waterproof membrane 18, and the adhesive composition 16. The roof deck 12 may be comprised of a number of materials including wood planks, plywood, cementitious wood fiber, gypsum, galvanized steel, painted steel, poured-in-place concrete, lightweight concrete, and precast concrete. Preferably, the roof deck comprises concrete.

15 Insulation 14 may be rigid board insulation including wood fiber, fiberglass, isocyanurate, expanded polystyrene, extruded polystyrene, Perlite, gypsum and Dens-Deck®.

To secure roofing insulation to the roof deck, all surfaces should be free of dust, dirt, oils, water and other contaminants. A conventional primer may be applied, 20 if desired. The adhesive is then preferably applied directly to the roof deck substrate using a ribbon pattern with  $\frac{1}{2}$ " to  $\frac{3}{4}$ " (1.3 cm to 1.9 cm) round beads to achieve proper coverage rates. For example, for concrete and treated plywood roof decks, 1 gallon per 100 square feet is preferred, while 2 gallons per 100 square feet (7.6 liters per 9.3 square meters) is desired in a reroofing application, where the adhesive is applied to a built up roof. We have found that four beads  $\frac{1}{2}$ " by 4' (1.3 cm to 1.2 meters) per 4' x 4' (1.2 m x 1.2 m) area yields a coverage rate equivalent to 1 gallon 25 per 100 square feet (3.79 liters per 9.3 square meters). Five beads  $\frac{3}{4}$ " by 4' (1.9 cm to 1.2 meters) per 4' x 4' (1.2m x 1.2 m) area yields coverage rate equivalent to 2 gallons per 100 square feet. The adhesive can be applied directly from the 30 packaging, using a ladle, or by using a multi-port adhesive applicator available from Industrial Metal Fabricators, Inc. Once applied, the insulation board may be immediately set in the wet adhesive. The adhesive cures fully within about 24 hours.

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The cure rate will be dependent on atmospheric temperature, relative humidity and moisture within the substrates.

It should be appreciated that while the adhesive composition of the present invention has been described for use in adhering roofing insulation to a roof deck, the 5 composition may also be used in coating applications and in caulking and sealing applications. The adhesive composition strongly adheres alone or with the aid of a primer to a wide variety of substrates such as glass, porcelain, metals and polymeric materials and the like, making it especially suited for various types of caulking, adhesive and laminating applications.

10 In order that the invention may be more readily understood, reference is made to the following examples which are intended to illustrate the invention, but not limit the scope thereof.

Example 1

An adhesive composition was prepared in accordance with the present 15 invention by mixing 54.66% by weight RT-7 coal tar, commercially available from AlliedSignal, Inc., 19.52% by weight diundecyl phthalate, and 2.15% by weight Isonate 50-OP (MDI) available from Dow Plastics. The components were mixed until homogeneous, for about 5 to 15 minutes. To this mixture was then added 0.007% by weight DABCO BL-19, commercially available from Air Products. The components 20 were again mixed until homogeneous, about 5 to 15 minutes and then continuously mixed under vacuum (minimum 20 inches Hg gauge (508 mm Hg gauge)) until the moisture level of the batch was about 400 parts per million, for about 50 to 70 minutes. A silylated polymer prepared in accordance with the present invention was then added (23.43% by weight) and mixed until homogeneous, about 5 to 15 minutes. 25 To this mixture was then added 0.12% by weight METACURE T-1 (available from Air Products) and 0.12% by weight Polycat DBU (available from Air Products). The components were mixed until homogeneous, about 5 to 15 minutes, and then continuously mixed under vacuum (minimum 20 inches Hg gauge) for 30 to 60 minutes.

Example 2

The adhesive composition of the present invention (designated as A) was applied to concrete and used to secure two types of roofing insulation. The adhesive strength was tested using a simulated wind uplift pull test on 2 ft. x 2 ft. (.6 m x .6 m) pieces of insulation secured to concrete. The test was conducted in accordance with approval standards set by Factory Mutual Research Corporation (FMRC).

The test results are shown below in Table 1 along with test results obtained from another commercially available product designated as B. Product B is an insulation adhesive formulation based on 15.0 to 40.0 parts by weight of a conventional polyurethane polymer and from 25.0 to 50.0 parts by weight asphalt.

Table 1

	A	B
	Isocyanurate insulation failed at 285 psf (139 g/cm <sup>2</sup> ) meets class I-315	meets class I-240
15	Wood fiber board insulation failed at 225 psf (109 g/cm <sup>2</sup> ) meets class I-240	meets class I-165

The classes indicated refer to ratings based on wind uplift pressure. The higher the rating, the higher the wind uplift pressure/wind speed the adhesive can withstand.

As can be seen, the adhesive strength of the adhesive composition A of the present invention is higher than that of B. It should be noted that adhesive A withstood the force applied; the failure was due to the substrate being torn apart.

Adhesive compositions A and B were further tested to compare adhesive strength immediately after application. Adhesive samples of A and B were applied to a series of 4" by 6" (10.1 cm x 15.2 cm) pieces of metal decking which were immediately adhered to 4" by 6" (10.1 cm x 15.2 cm) pieces of plywood. The adhered samples were allowed to age for the times noted in Table 2 below. The aging conditions were 72°F (22°C) and 30% relative humidity. The samples were pulled on an Instron testing machine at a rate of 2 inches/minute. The results are shown below:

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Table 2

## Adhesive Strength

Dwell Time	A	B
5	15 minutes	2.3 psi (162 g/cm <sup>2</sup> )
	30 minutes	4.5 psi (316 g/cm <sup>2</sup> )
	1 hour	8.6 psi (605 g/cm <sup>2</sup> )
	2 hours	10.2 psi (717 g/cm <sup>2</sup> )
	4 hours	13.8 psi (970 g/cm <sup>2</sup> )

As can be seen, the initial strength of the adhesive A of the present invention is  
 10 much higher than that of adhesive B throughout the first 4 hours after application.  
 This is believed to be due the faster rate of cure of the adhesive of the present  
 invention. The higher initial adhesive strength and faster rate of cure provides a more  
 secure bond and reduces the chances that the insulation will be moved or dislodged  
 during installation.

15 It will be obvious to those skilled in the art that various changes may be made  
 without departing from the scope of the invention which is not considered limited to  
 what is described in the specification.

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CLAIMS

1. A one-component, moisture-curable adhesive composition comprising:
  - a) a silylated polymer selected from the group consisting of silylated polyurethanes and silylated polyethers; and
  - 5 b) an extender selected from the group consisting of coal tar, aromatic oils, and hydrocarbon resins.
2. The adhesive composition of claim 1 wherein said extender comprises coal tar.
3. The adhesive composition of claim 1 further including a plasticizer selected  
10 from the group consisting of phthalates, mellitates, benzoates, phosphates, butyrates, polymeric esters, adipates, sebacates, aromatic and cyclic oils, epoxidized oils, and solvents.
4. The adhesive composition of claim 3 wherein said plasticizer comprises diundecyl phthalate.
- 15 5. The adhesive composition of claim 1 further comprising a moisture scavenger.
6. The adhesive composition of claim 5 wherein said moisture scavenger comprises 4,4,-diphenylmethane diisocyanate.
7. The adhesive composition of claim 1 further including at least one catalyst selected from the group consisting of dibutyl tin diacetate, 1,8-diaza-bicyclo (5.4.0)  
20 undecene-7, (bis(2-dimethylaminoethyl)ether, and combinations thereof.

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8. An adhesive composition for securing insulation to a roof deck comprising:
  - a) from about 20 to 50% by weight of a silylated polymer;
  - b) from about 30 to 75% by weight of an extender;
  - c) from about 2 to 25% by weight of a plasticizer; and
  - d) from about 0.5 to 10% by weight of a moisture scavenger
- 5 9. The adhesive composition of claim 8 wherein said silylated polymer is selected from the group consisting of silylated polyurethanes and silylated polyethers.
- 10 10. The adhesive composition of claim 8 wherein said extender comprises coal tar.
- 10 11. The adhesive composition of claim 8 wherein said plasticizer is selected from the group consisting of phthalates, mellitates, benzoates, phosphates, butyrates, polymeric esters, adipates, sebacates, aromatic and cyclic oils, epoxidized oils, and solvents.
- 15 12. The adhesive composition of claim 8 wherein said plasticizer comprises diundecyl phthalate.
13. The adhesive composition of claim 8 wherein said moisture scavenger comprises 4,4,-diphenylmethane diisocyanate.
- 20 14. The adhesive composition of claim 8 further including at least one catalyst selected from the group consisting of dibutyl tin diacetate, 1,8-diaza-bicyclo (5.4.0) undecene-7, (bis(2-dimethylaminoethyl)ether, and combinations thereof.
- 25 15. A roof system comprising roofing insulation secured to a roof deck substrate, wherein said insulation is secured by an adhesive composition comprising a silylated polymer selected from the group consisting of silylated polyurethanes and silylated polyethers and an extender selected from the group consisting of coal tar, aromatic oils, and hydrocarbon resins.

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16. A method of adhering roofing insulation to a roof deck comprising:

5      a) applying an adhesive composition to a roof deck substrate, said adhesive composition comprising a silylated polymer selected from the group consisting of silylated polyurethanes and silylated polyethers and an extender selected from the group consisting of coal tar, aromatic oils, and hydrocarbon resins;

      b) placing said insulation in contact with said adhesive composition on said roof deck substrate; and

      c) curing said adhesive composition.

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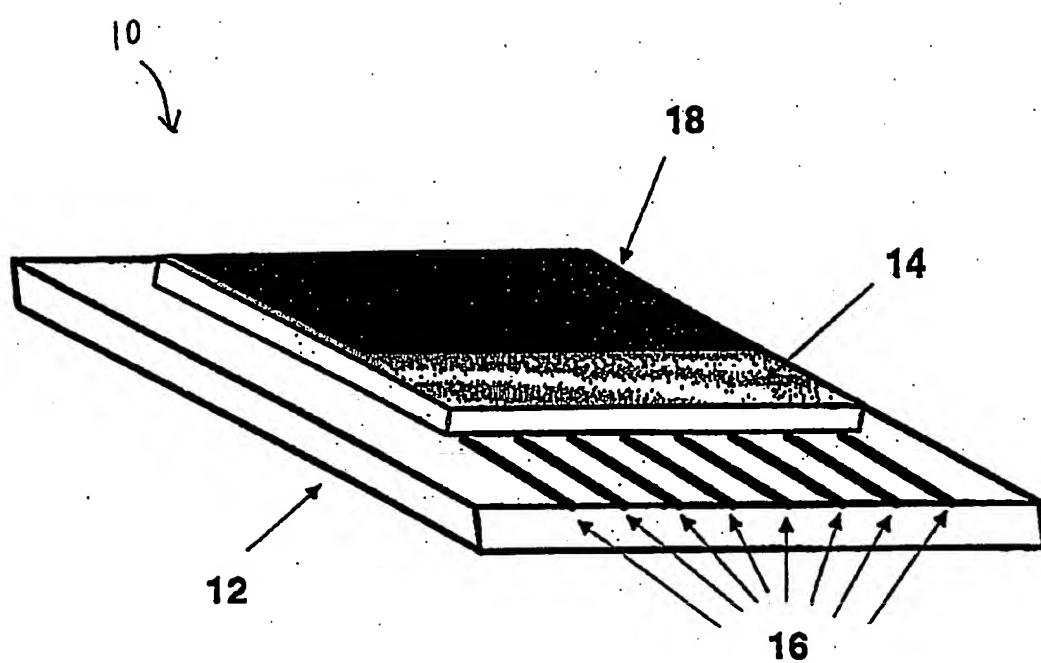


FIG. 1

# INTERNATIONAL SEARCH REPORT

International Application No.  
PCT/US 99/30941

**A. CLASSIFICATION OF SUBJECT MATTER**  
IPC 7 C08G65/336 C08G18/10 C08G18/08 C09J201/10

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)  
IPC 7 C08G C09J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 336 431 A (KANEKA FUCHI CHEMICAL INDUSTRY) 11 October 1989 (1989-10-11) page 2, line 50 -page 8, line 5; claims 1-8 —	1
X	DATABASE WPI Week 9538, 1995 Derwent Publications Ltd., London, GB; AN 290656 XP002135456 "adhesive composition" & JP 07 188641 A (KONISHI), 25 July 1995 (1995-07-25) abstract —	1 —/—

Further documents are listed in the continuation of box C.

Patent family members are listed in annex.

\* Special categories of cited documents :

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Date of the actual completion of the International search	Date of mailing of the International search report
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# INTERNATIONAL SEARCH REPORT

Intern. Appl. No.  
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## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	EP 0 844 282 A (KANEKA FUCHI CHEMICAL INDUSTRY) 27 May 1998 (1998-05-27) page 2, line 29 -page 7, line 2; claims 1-8	1,3,5,7
Y	EP 0 819 749 A (SIMSON) 21 January 1998 (1998-01-21) page 2, line 20 -page 3, line 52; claims 1-3,13,14	1,3,5,7

**INTERNATIONAL SEARCH REPORT**

Information on patent family members

International Application No	
PCT/US 99/30941	

Patent document cited in search report	Publication date	Patent family member(s)		Publication date
EP 336431 A	11-10-1989	AU 3258689 A CA 1338924 A DE 68920614 D DE 68920614 T ES 2070136 T JP 2028277 A JP 2902667 B US 5631082 A		12-10-1989 25-02-1997 02-03-1995 06-07-1995 01-06-1995 30-01-1990 07-06-1999 20-05-1997
JP 7188641 A	25-07-1995	NONE		
EP 844282 A	27-05-1998	JP 9053010 A JP 9053011 A AU 6669496 A CA 2229048 A WO 9706212 A		25-02-1997 25-02-1997 05-03-1997 20-02-1997 20-02-1997
EP 819749 A	21-01-1998	GB 2315491 A GB 2327425 A		04-02-1998 27-01-1999